BERKELEY LAB

LUX - Linac-based Ultrafast X-ray source

A Recirculating Linac/Laser-based Femtosecond Facility for Ultrafast Science

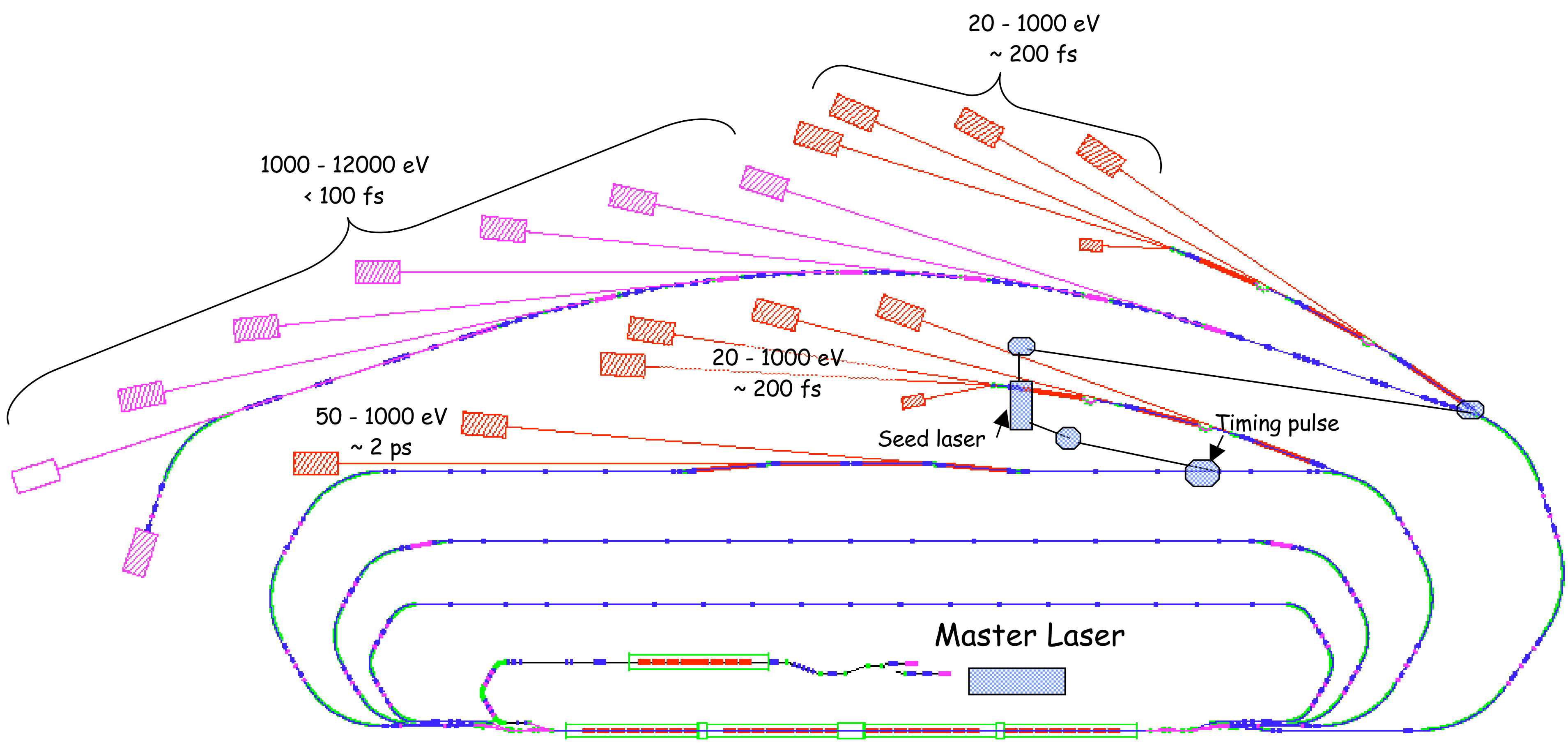
Time-resolved experiments

- Science and the multiple relationships between time, spectroscopy, and diffraction

- By combining diffraction and spectroscopy (nuclear positions and electronic, chemical or structural probes), outstanding new science will be achieved in the x-ray regime.

- Time dynamics parameters have not been exploited in the X-ray, mostly due to lack of sources.

Overall scope of the project: EUV, soft x-ray, and hard x-ray components

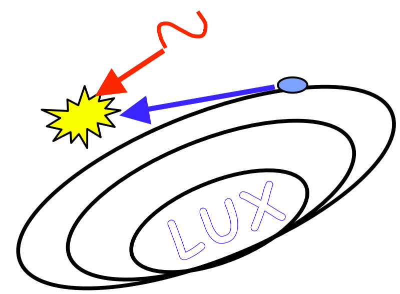


Ultrashort pulse duration, synchronization, and tunability - timing with femtosecond lasers for wide array of experiments

Specifications of the Facility

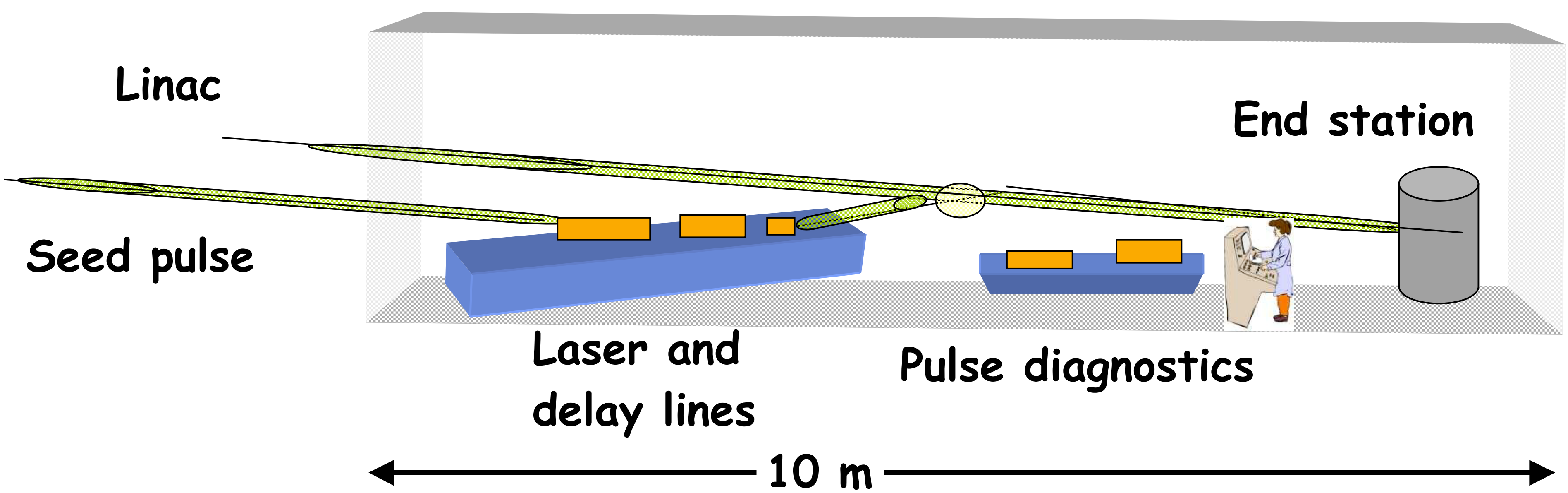
An ultrafast x-ray science user facility addressing scientific needs in Physics, Chemistry and Biology

- National user facility
  - Recirculating linac-based light source
  - multiple beamlines
  - laser-coupled end stations
- Repetition rate 10 kHz
- Synchronization ~ 10's fs
- Pulse durations 50-200 fs or less
- Polarization fully variable
- Broad photon range ~ 0.02 - 12 keV
- Photons per pulse 10^7 hard x-ray, 10^8-10^{12} soft x-ray



Typical End Station Layout

Precisely timed laser and linac pulses



Tunable laser systems designed for specific experiments, repetition rate, energies

Parameters that matter most to ultrafast scientists

Searching for weak dynamically changing signals in the midst of large time-invariant signals

- Timing

  most experiments performed by initiating a time evolving process with another laser or x-ray pulse - tight time synchronization is expected

  - 10's of fs for seeded, 50 fs for hard x-rays

- Pulse-to-pulse stability

  essential since only a small fraction of the molecules or materials are excited

  - Expectation of 3rd generation 0.1% stability, Real time subtraction - pump on/pump off

- Bandwidth and chirp

  BW a minimum, without violating transform limit, to isolate spectral shifts, chirp to correlate energy with time in new ways

  - Core level shifts of 0.1-0.5 eV typical, NEXAFS ≤0.1 eV desirable

Con't

Con't

Ultrafast scientists' needs

- Tunability
  - Spectroscopy demands tuning to near edge transitions
- Polarization
  - Complete rhc and lhc components needed for polarization blocking and dichroism experiments
- Repetition rate
  - High repetition rates desirable for samples that can be refreshed, low damage, as high as conventional electronics
- Pulse duration
  - 50-200 fs for many processes, 10 fs for future applications, 100 attosecond beyond
- Pulse energies
  - Sufficient pulse energies to obtain photoemission signals, absorption contrast changes, without sample damage
- Coherence
  - Spatial and temporal, speckle experiments

Con't

Ultrafast scientists' needs

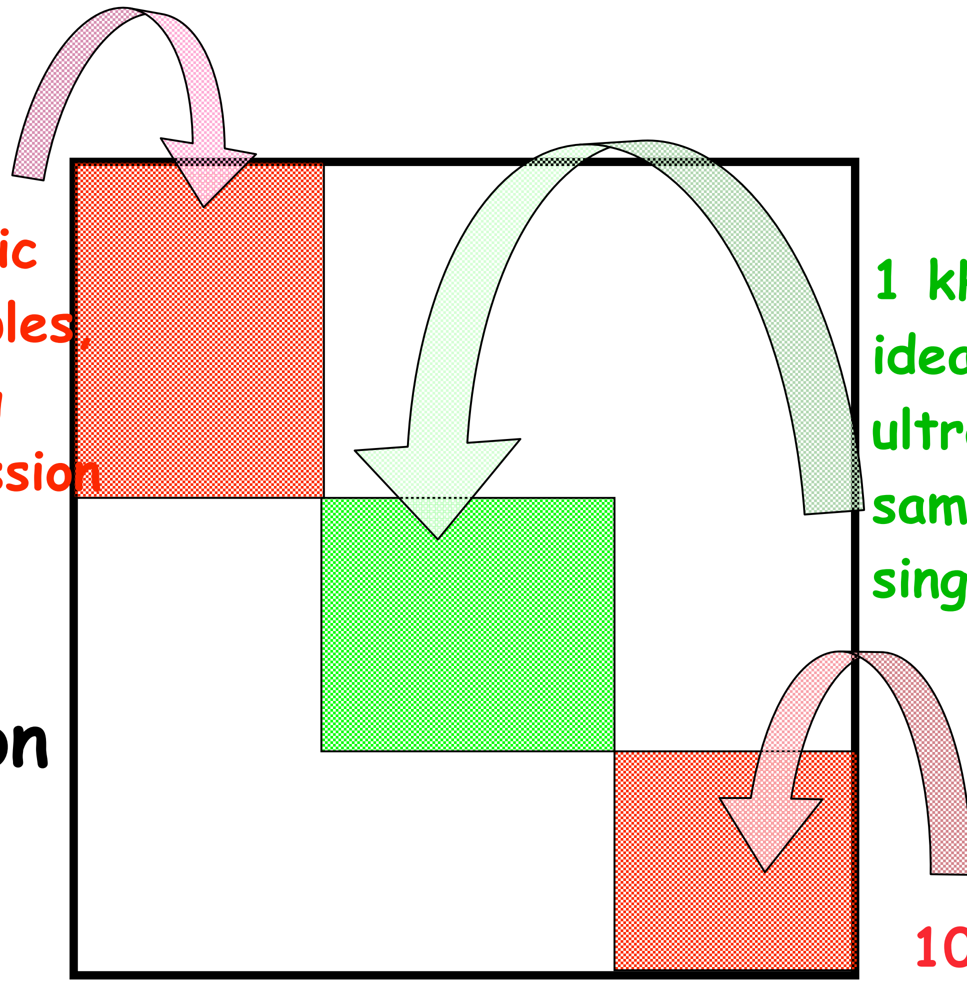
- Focusability
  - Near-diffraction limit for seeded systems, 10's nm at 1 keV
- Power density
  - 10^{15} W/cm^2 readily achievable
- Trade off between power density and repetition rate
  - Maintain linear probing for many experiments
  - Multiphoton versus single photon

cf. Wabnitz et al, Nature, 420, 482 (2002) - extreme high order multiphoton processes in Xe clusters with 100 fs FEL pulses at 98 nm and 7 x 10^{13} W/cm^2

LINAC design gives best opportunity to achieve experimental and facility goals

Repetition rate vs. Energy

MHz-GHz, nJs, limited by acoustic velocities in samples, good for counting expt's, photoemission imaging



1 kHz-100kHz, □Js ideal match to ultrafast lasers and sample considerations, single photon regime

10-100Hz, mJs, limited by sample damage, many high field effects

↑ Repetition rate

Pulse energy →

Liquid Microjet Studies of Surface Structural Changes on Ultrafast Timescales

An example of an experiment that requires:

- Narrow Bandwidth
- Pulse-to-pulse stability
- Tunability

Liquid water molecules

- Probe O atom K edge at 530 eV
- Obtain structure and bonding of surface water molecules
- Sensitively probed by short escape depth of ions

New ultrafast experiments

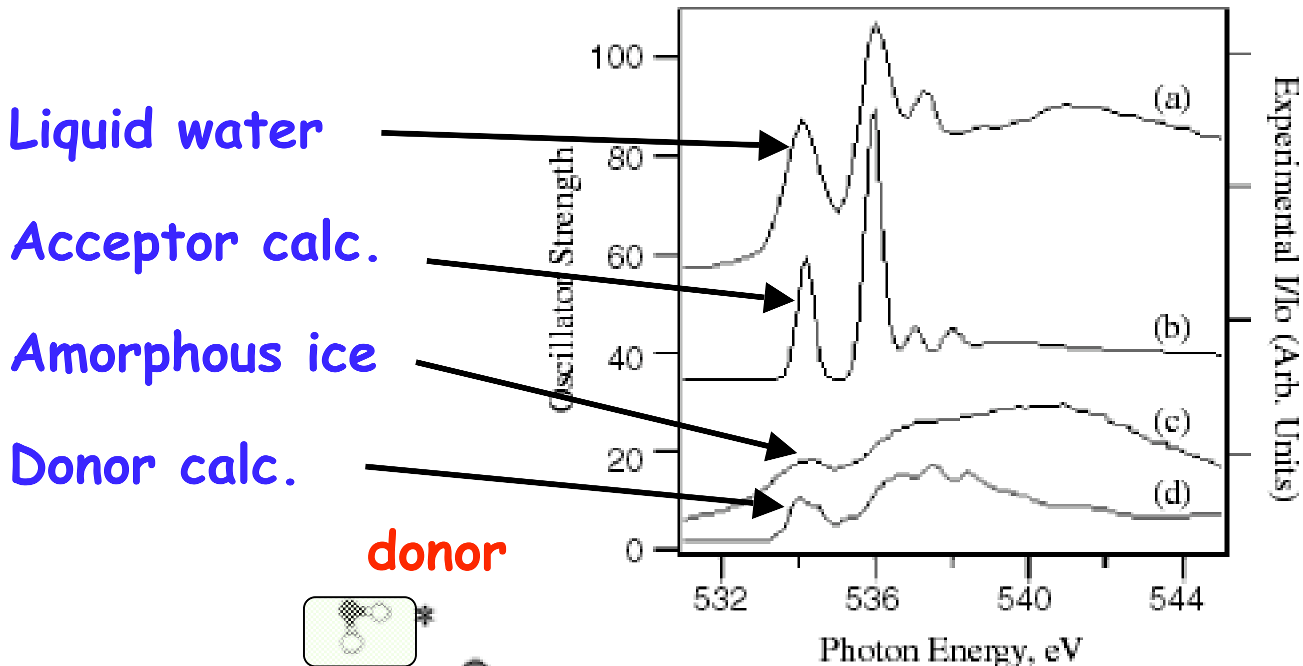
- Excite Surface molecules vibrationally and photolytically and observe surface structural changes, time domain interfacial studies, caging
- Hydrocarbon, salt and alcohol dopants segregate at surface, rich chemistry

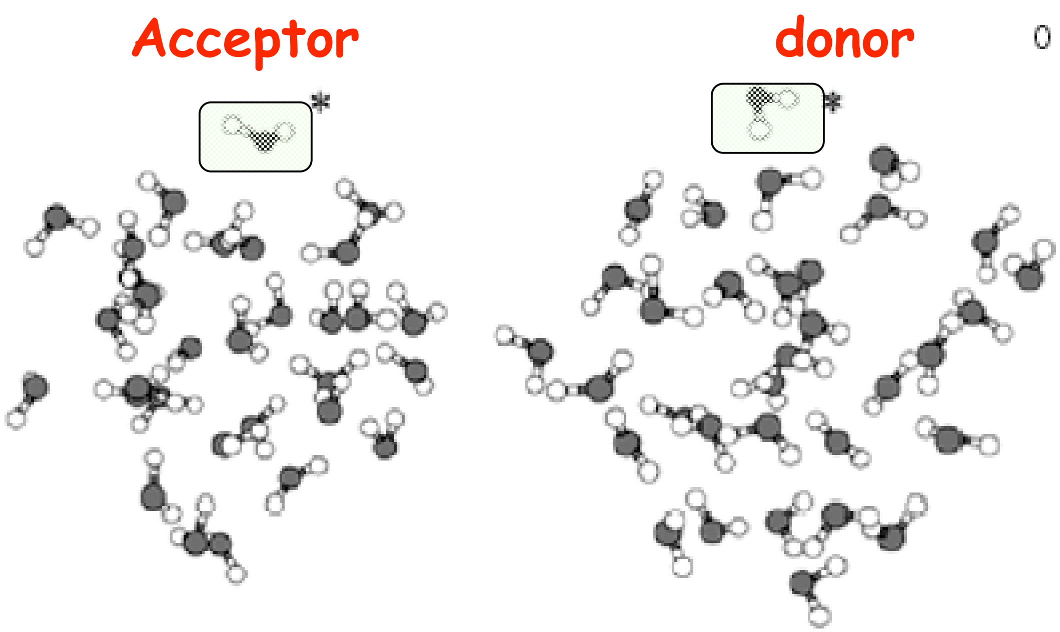
Advantages

- Complete sample regeneration
- Power densities limited primarily by space charge, affects imaging

Based on work of Saykally, et al.

Experimental NEXAFS of Liquid Water Surface





Hydrogen-bonded donor and acceptor

Acceptor structure fits better

Linac properties - liquid jet exp't

Assume 1-10% changes in surface structural bond orientations upon excitation (heating) or photolysis of species segregated at surface

Bandwidth: 0.1 eV achievable with external monochromator, t= 50 fs

Tunability: tunable in seconds, steps of 0.1 eV with seed laser (EUV sum freq output) and monochromator. Entire time map of 10 eV takes a few hours.

Integrated stability: pulse-to-pulse stability of 20% translates to 0.1% integrated stability in 4 second integration times at 10 kHz

Laser-on versus laser-off signals acquired on every other laser pulse with two multichannel scalers.

Phase Transitions

- **Phase transitions induced by fs or ps pulses**

VO_2 Monoclinic (insulator) □ Rutile (metal) transformation

first order phase transition

T=340 K or fs pulse excitation

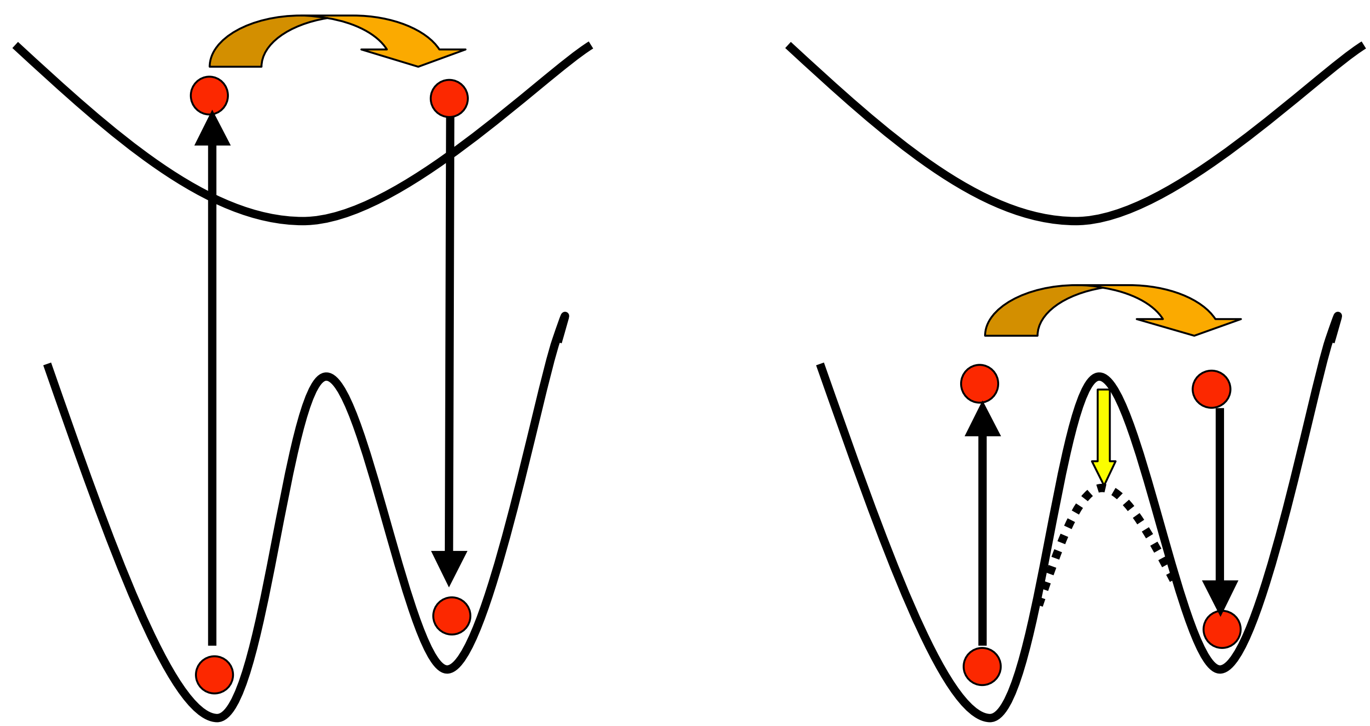
(Cavalleri et al. PRL,87, 237401 (2001))

Carrier excitation can directly alter the electronic potential surface or thermally-induced atom displacements can overcome the barrier or lower the barrier for the insulator-to-metal transition

Combine NEXAFS with Bragg with Fermi photoemission

Solid State Phase Transformations

Multiplicity of mechanisms for phase transitions

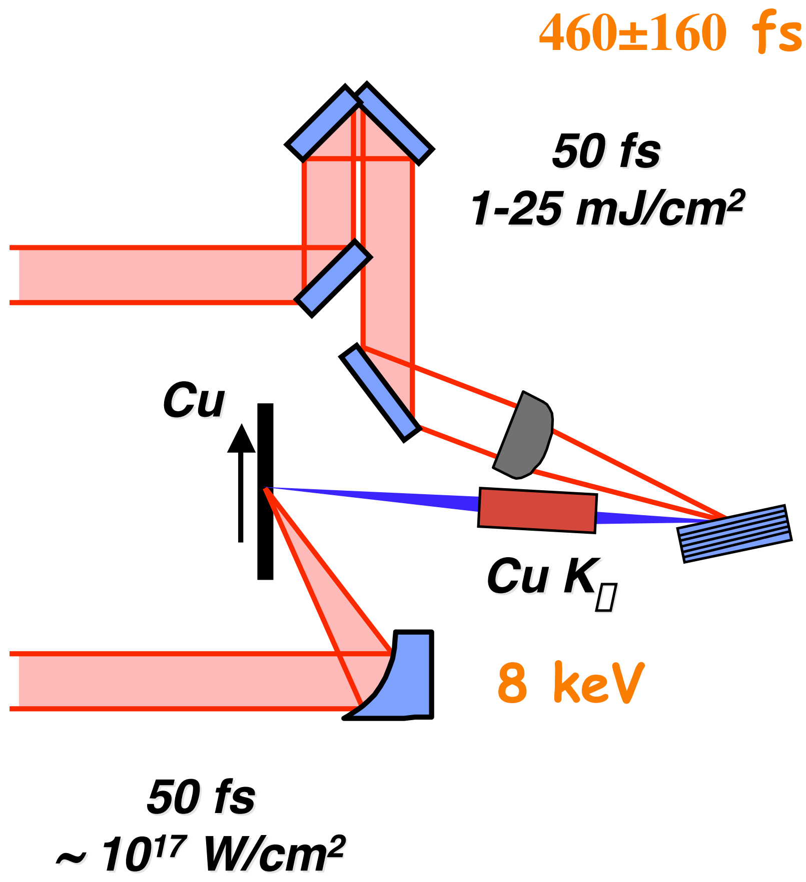


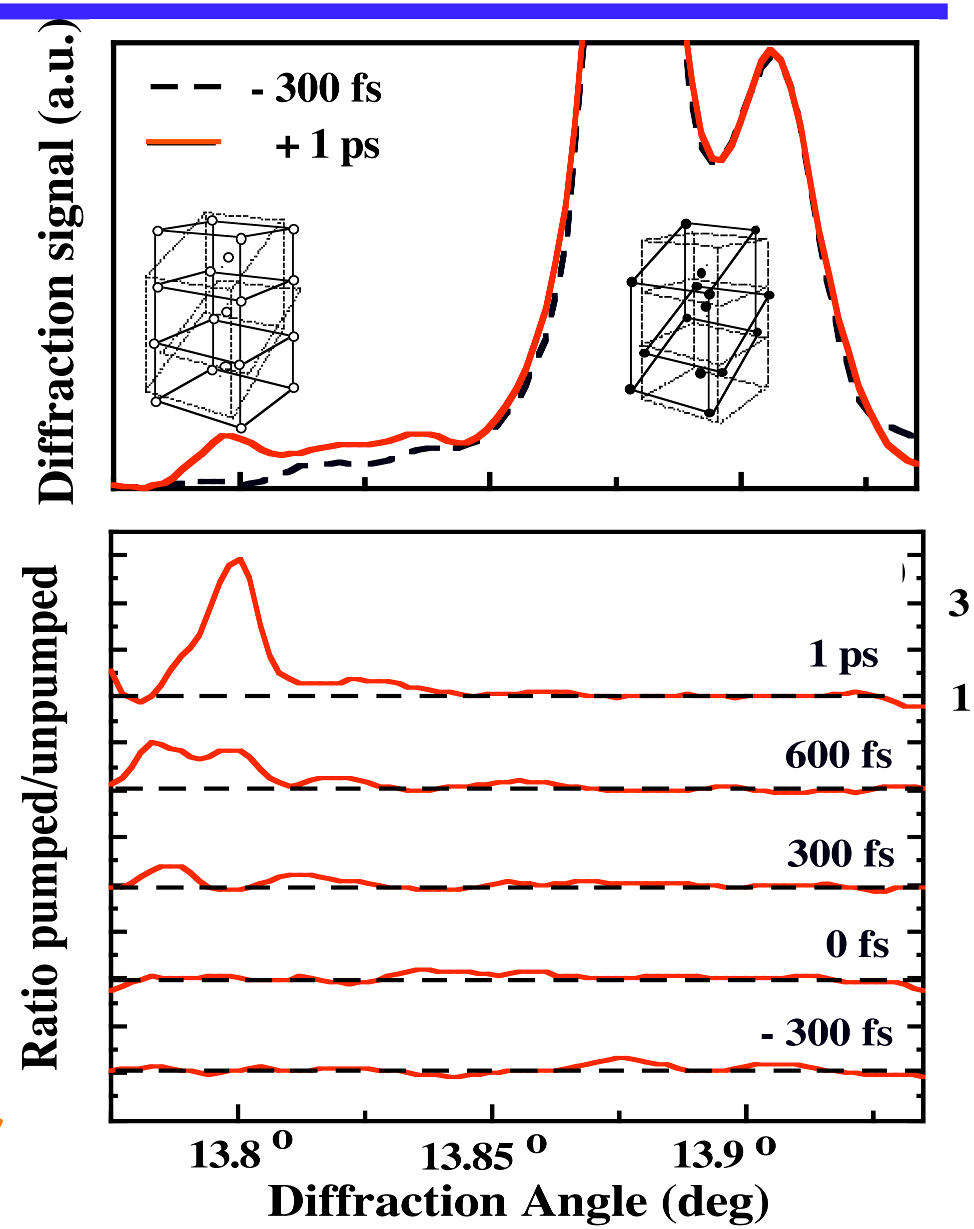
Phase change

Photoinduced

thermal activation

Bragg Diffraction





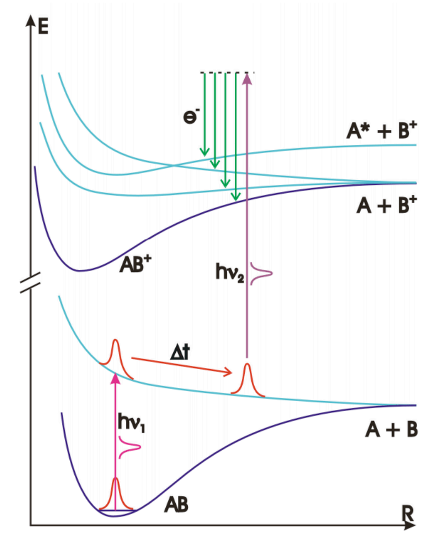
Only 2000 Cu K photons per pulse, 20 Hz, only topmost layers altered

fs-XPS of molecular transformations

Core level and valence shell electrons are a powerful means to characterize chemical environments

At least four new types of fs spectroscopies possible:

- dissociative state and bound excited state valence shell photoelectron spectroscopy (PES)
- dissociative state and bound excited state x-ray photoelectron spectroscopy (XPS)

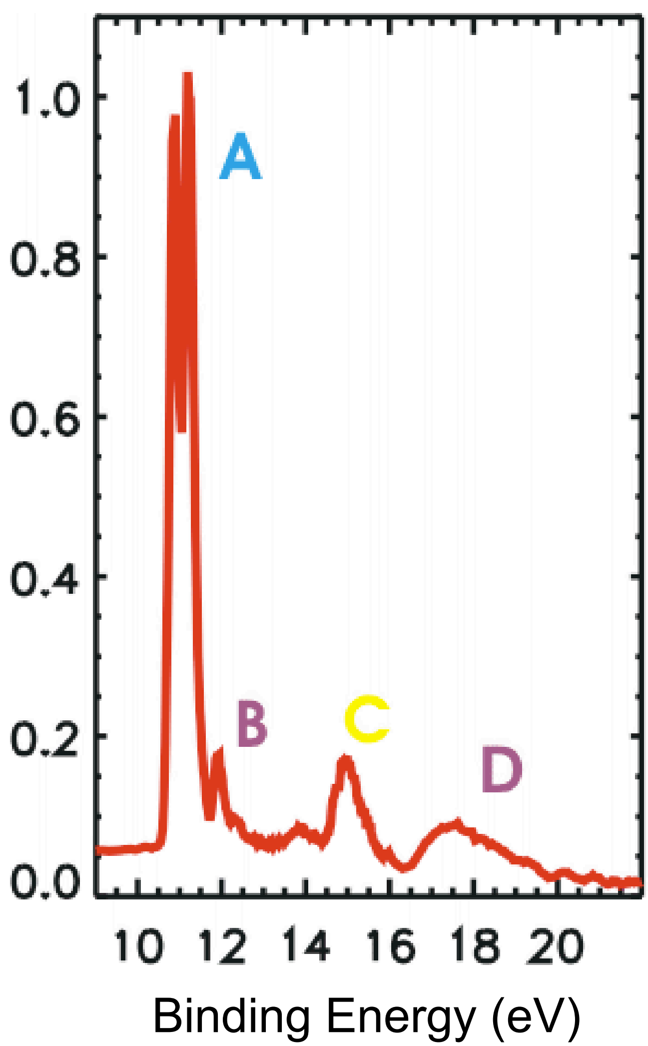


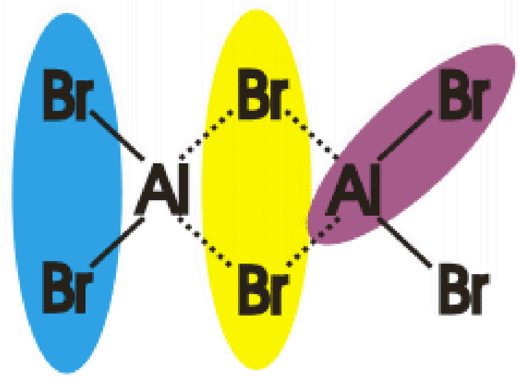
Valence and core electrons provide complementary information about bonding and atomic "chemical" environment

Address problems of femtosecond charge switching dynamics, XPS spectra of highly vibrationally excited molecules, excited state dynamics and potential surfaces

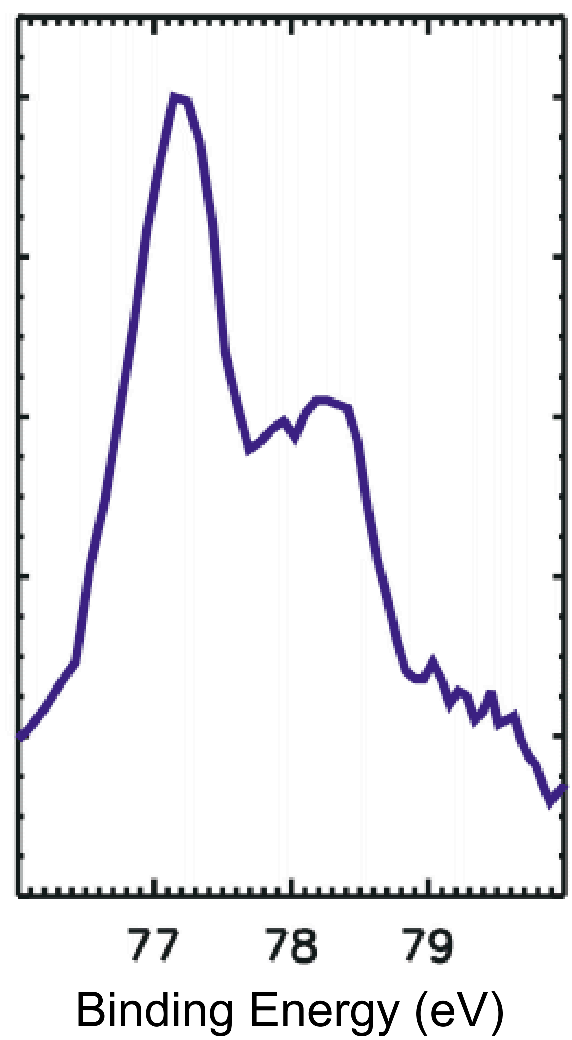
•Photoelectron Spectra of Al_2Br_6

17th harmonic

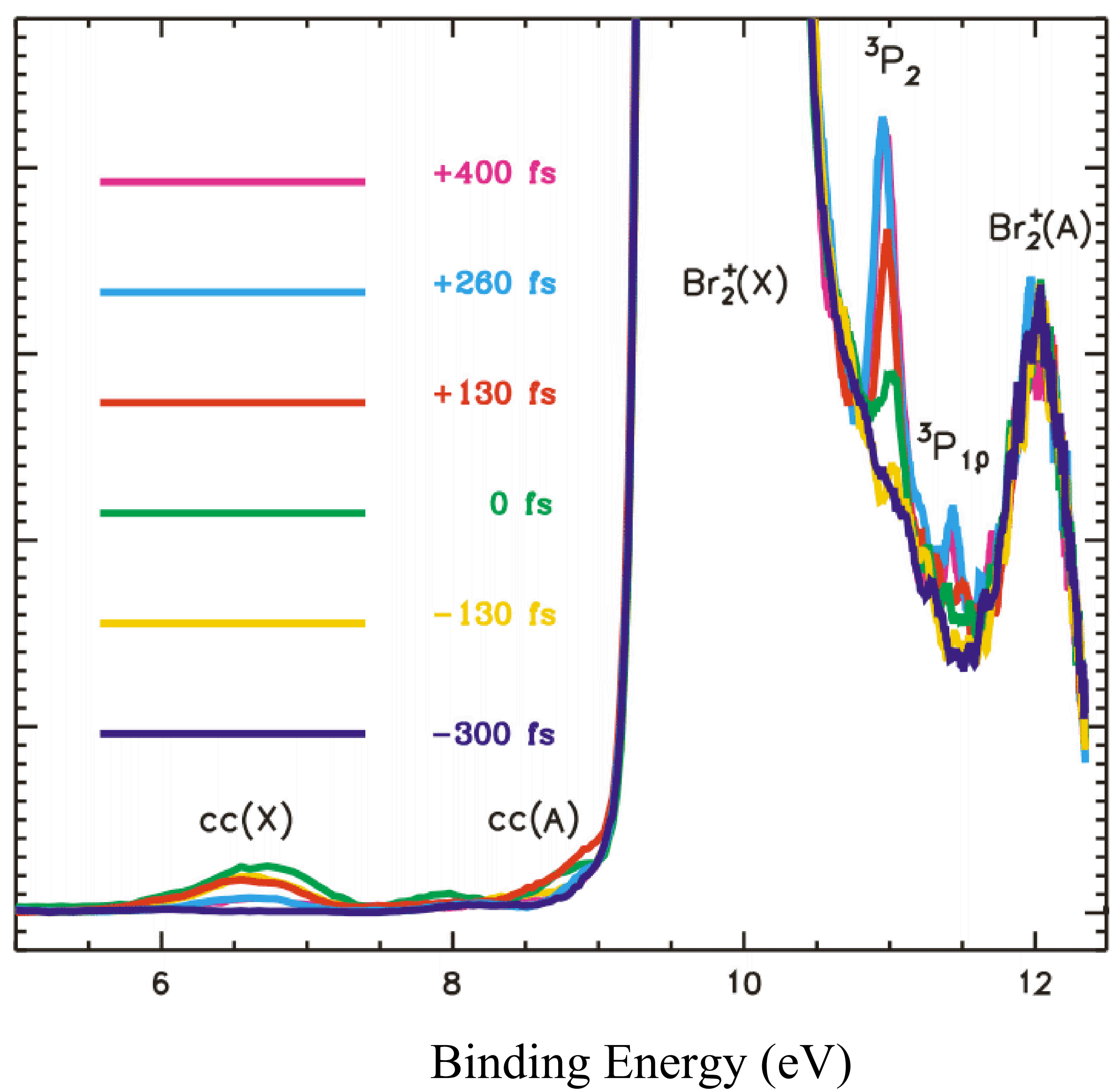




55th harmonic

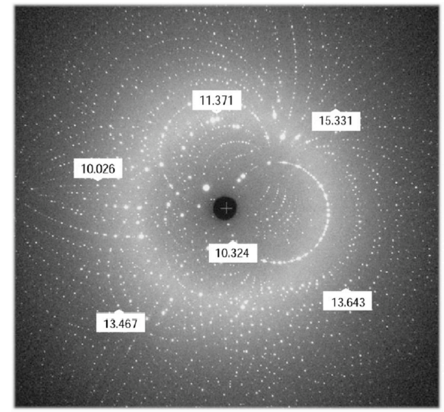


fs photodissociation of Br_2



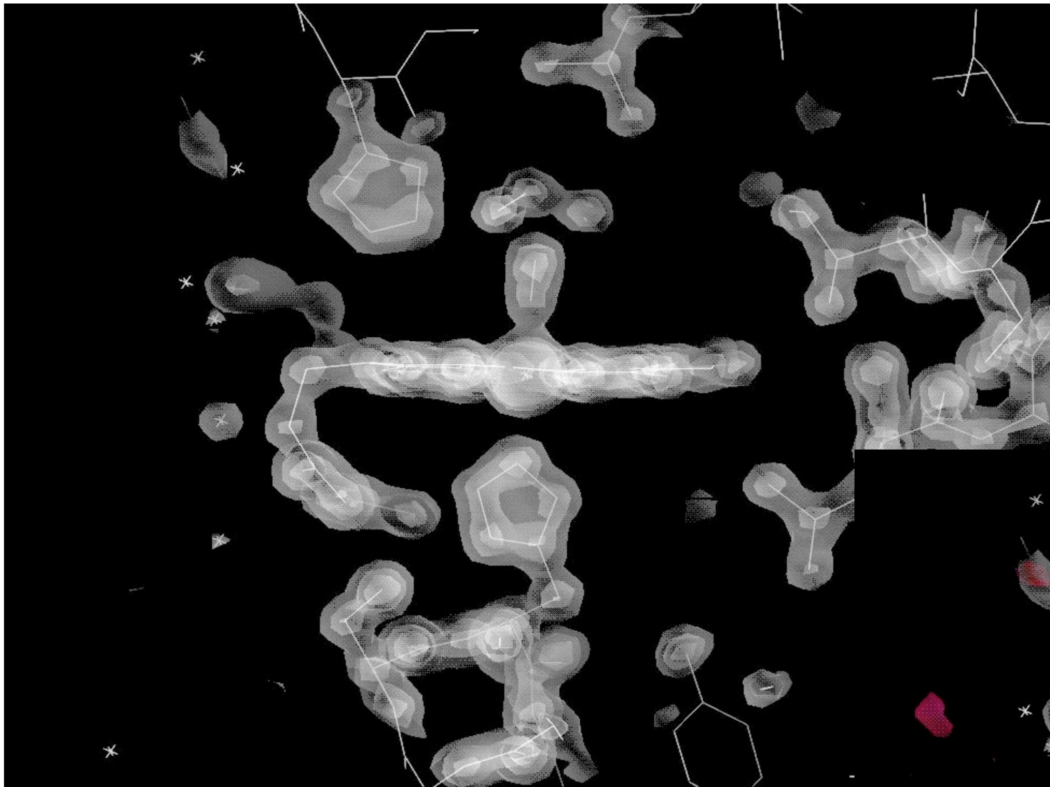
Biomolecule Crystallography

- Key time-resolved x-ray Laue diffraction experiments demonstrated at ESRF on photoactive systems, single crystal myoglobin and yellow protein
- Some biological processes inherently slow, others impressively fast, and these will become targets of investigation
- More systems will be developed and explored on ultrafast timescales as fs x-ray sources become available



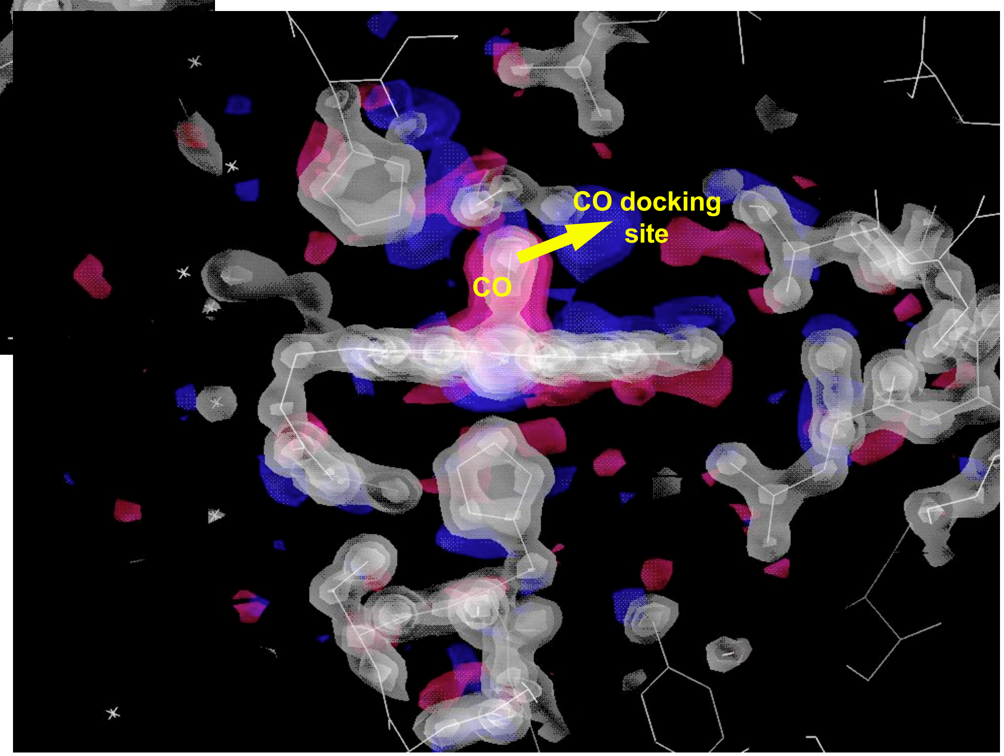
- Probe desirable photochemically active, reversible sites related to plant photochemistry and vision, as well as the time-resolved nonreversible photon and particle damage of biologically active materials, related to cancer and mutations

Biological Structural Studies



ESRF ID09 10^7 photon per pulse per 0.1% BW

LUX is comparable, with higher repetition rate



3 ns after optical pulse

Methods of Time-Resolved Crystallography

- Pump-probe experiments, optical excitation, x-ray Laue diffraction, 10 keV and higher x-rays
- Difference electron density maps
- Time dependent intermediate structures extracted at the data analysis stage (so-called analytical trapping, compared to chemical trapping or physical trapping by lowering temperature)
- Movies of transformations to follow an array of intermediates

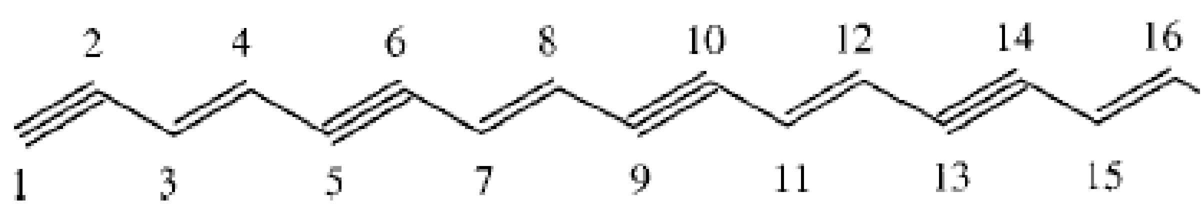
Novel limits of short pulse x-rays

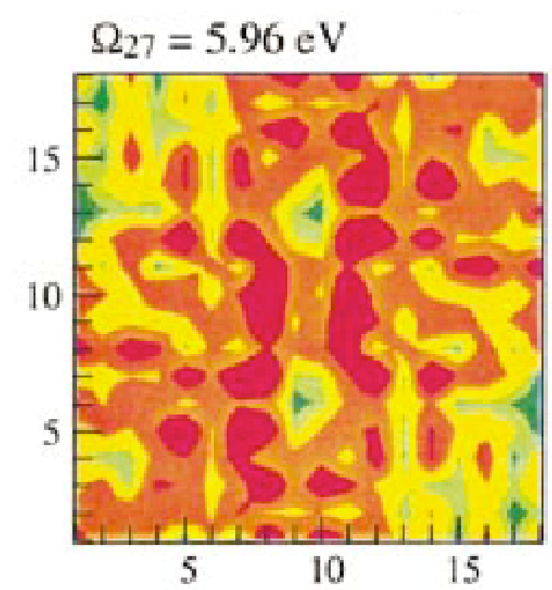
Time-resolved x-ray Raman spectroscopy

An inelastic Raman scattering of photons that reveals valence electron states

Novel probe of electronic and vibrational motions in time

Signals are shifted in energy a few eV from the large elastically scattered photon flux



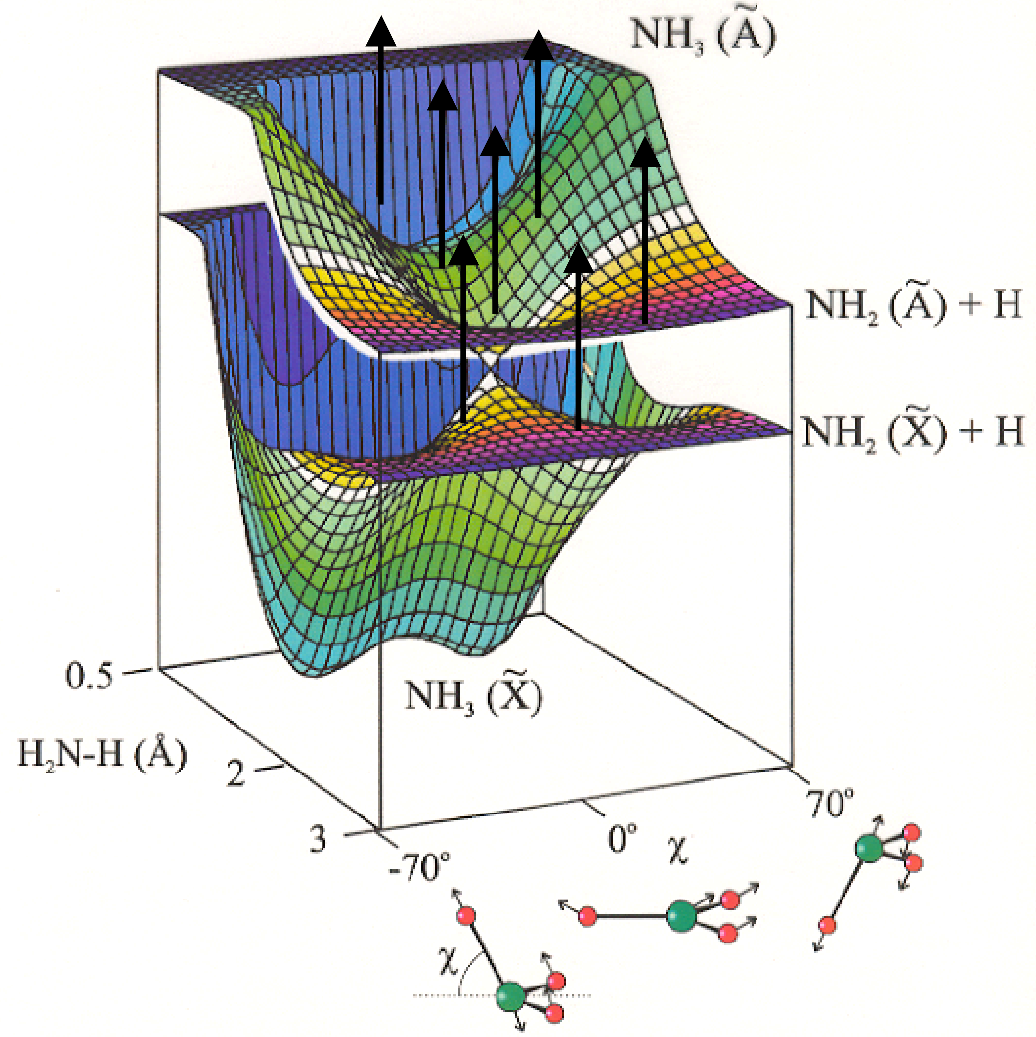


Tanaka, Volkov, Mukamel - polydiacetylene

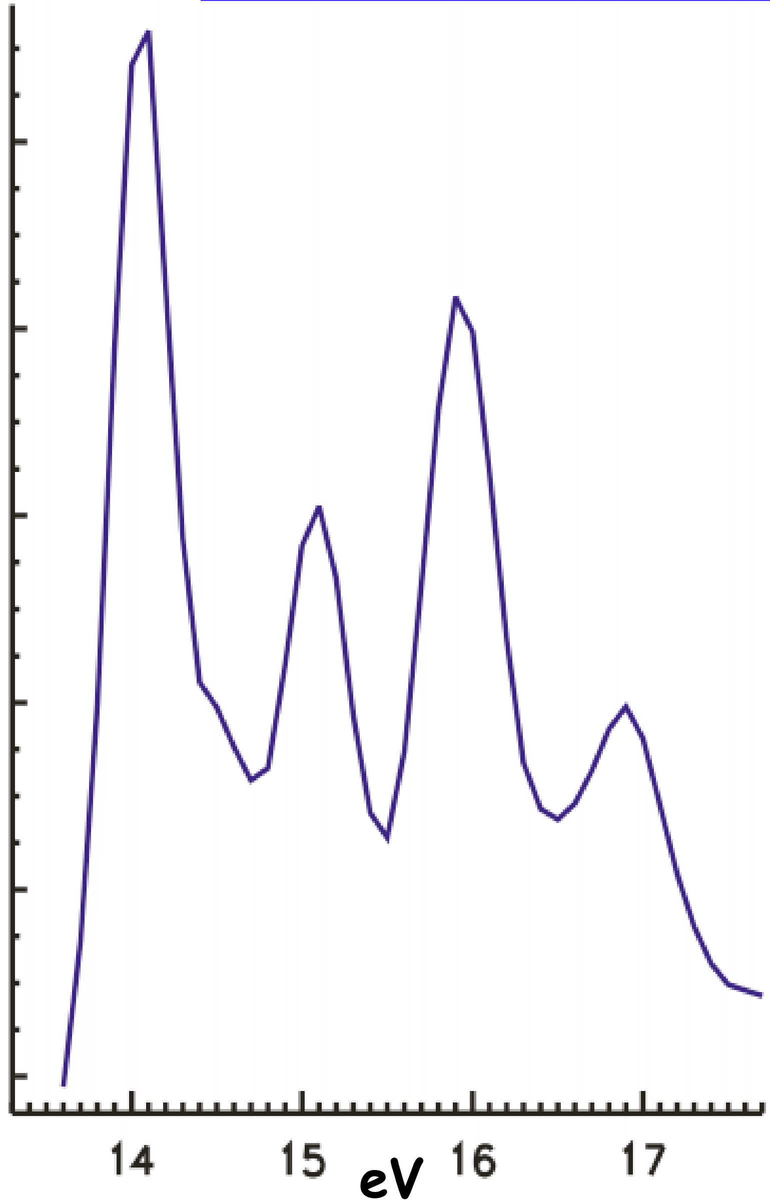
Transition density matrix for 27th excited state vs. carbon atom position

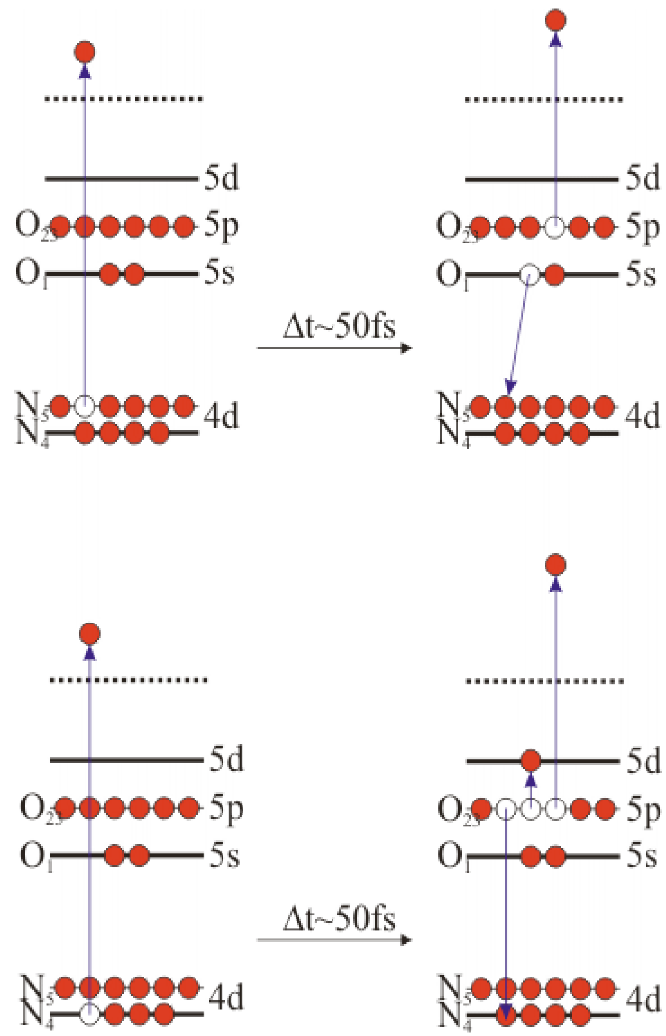
Atom specific probing of transition states in real time

Probing transition states in real time



- “Femtosecond” Auger spectroscopy

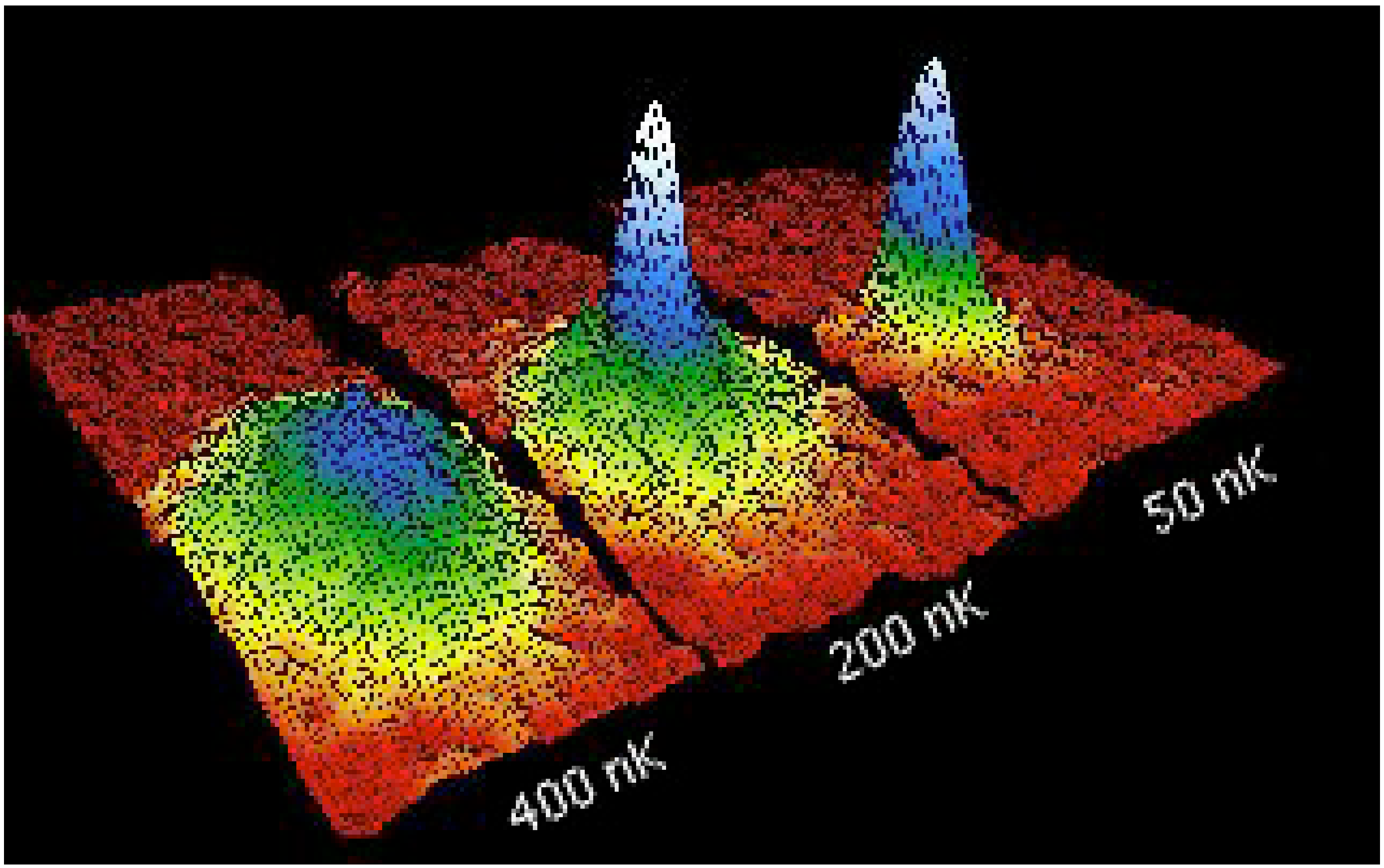




“dream” exp’ts on sub-fs timescales

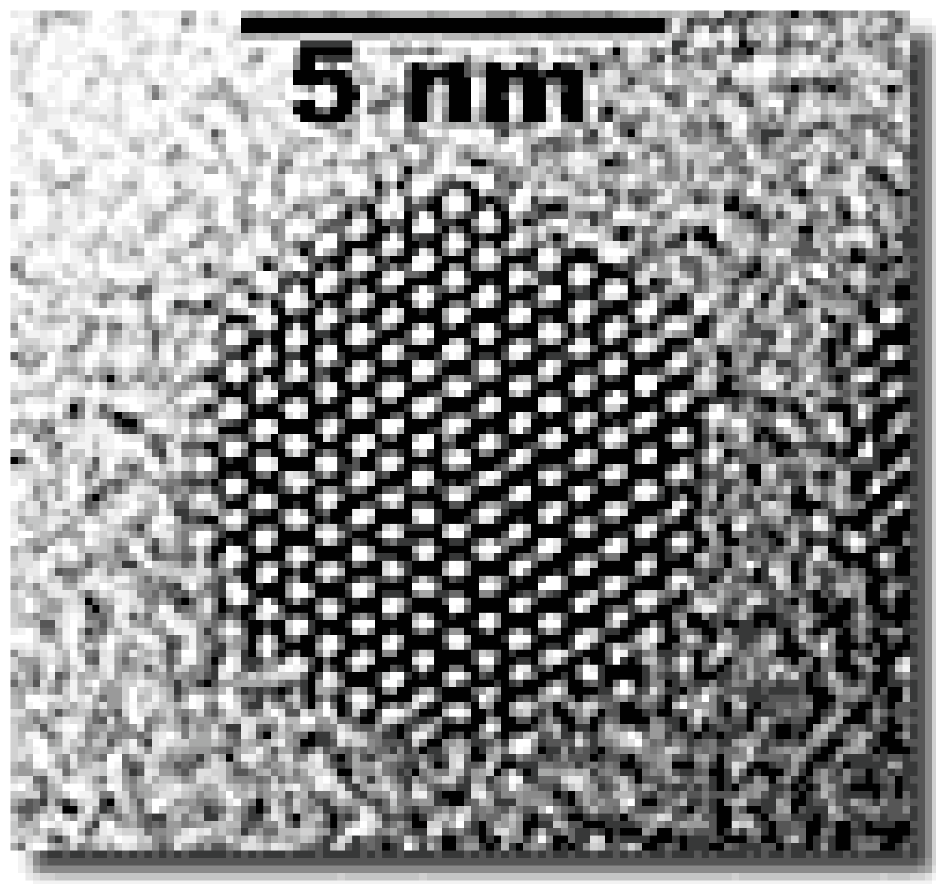
Ultrafast perturbations and Bose Einstein Condensates

Quantum mechanical, fragile states of matter, localized ordering and correlated interactions



Quantum Dots, Superposition States and Quantum Information

Semiconductor quantum dots - probing localized charge distributions, confinement, and exciton entanglement



The LUX Science Case

Why in Berkeley - strong groups in fs science in the Bay Area - strong coupling to laser community: Chemla, Falcone, Fleming, C. Harris, Leone, Orenstein, Schoenlein, Shank, Fayer, S. Harris, Livermore laser expertise, proximity to LCLS.

- the ALS community of scientists and engineers
-excellent user base for both soft and hard x-rays
-array of spectroscopy and microscopy expertise
-outstanding accelerator design team

The fs Linac Science Case

Why unique and important - National and international user base - young scientists interested in ultrafast processes, tremendous grass roots efforts growing in ultrafast x-ray science - answer critical questions with national need

-recirculating linac design is fundamentally a different source from SASE process

-an excellent,highly refined platform for a user facility

-allows major national thrust for time-dynamics investigations in the x-ray

-timing and synchronization, matched to laser excitation sources, laser seeding are central concepts for success